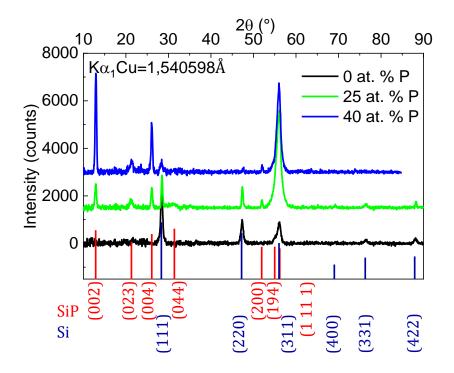
## Supplementary Information Thin Films of SiP Lamellar Alloys: a First Step Towards 2D SiP

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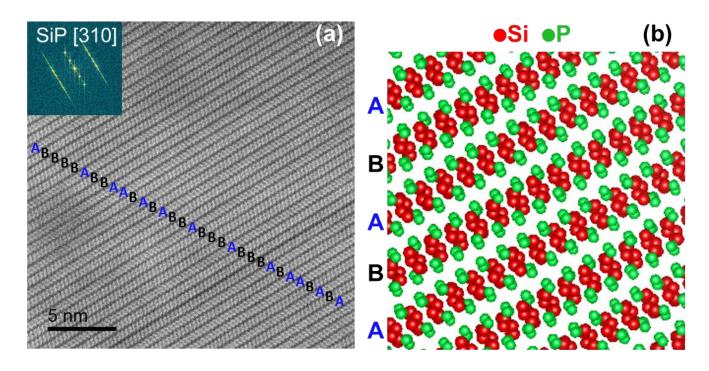
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Figure S1 shows grazing incidence X-ray diffraction patterns for Si:P thin films containing 0 at.% of P (black solid line, reference sample), 25 at.% of P (green solid line) and 40 at. % of P (blue solid line) after annealing at 1100°C for 5 minutes.



<u>Figure S1</u>: Grazing incidence X-ray diffraction patterns for Si:P thin films containing 0 at.% of P (black solid line), 25 at. % of P (green solid line) and 40 at. % of P (blue solid line) after annealing at 1100°C for 5 minutes. The blue and red vertical lines indicate the diffraction peaks associated with Si and orthorhombic SiP, respectively.

The incidence angle was fixed at  $2^{\circ}$  to minimize the contribution from the Si(001) substrate. In this case, Si:P thin films, which are not overgrown with SiO<sub>2</sub>, are considered. For the reference sample, one can clearly identify several peaks at 28.4°; 47.3°; 56.1°; 69.1°; 76.4° and 88° which correspond to the (111), (220), (311), (400), (331) and (422) reflections of crystalline Si (JCPDS file 00-026-1481). These reflections originate from Si present in the thin film. The relative intensities suggest a random crystallization of the Si grains in the films without particular texturing. For the samples containing 25 at.% of P and 40 at. % of P, the Si-related peaks are still present but additional peaks can be identified at 13°; 21.4°; 26.2°; 31.5°, 52.1°, 54.9° and 56°. The latter correspond to the (002), (023), (004), (044), (200), (194) and (1 11 1) directions of orthorhombic SiP (JCPDS file 01-073-1251). X-ray diffraction measurements thus confirm the presence of both Si and SiP areas in our thin films.



<u>Figure S2</u>: STEM-HAADF image of a SiP grain oriented along the [310] zone axis. The corresponding Fourier transform is shown in the inset. The atomic structure along the [310] zone axis direction is shown in (b).The normal structure can be described as the repeating stacking of A- type and B- type layers. Staking faults are clearly visible in the grain.

Figure S2 shows a STEM-HAADF image of a SiP grain oriented along the [310] zone axis. One can observe a chevron-like shape alternating from one atomic row to the next one. To get a better insight into the microstructure, we have taken a FFT of the SiP grain (see inset of fig. S2(a)). A good agreement is found when assuming an orthorhombic structure for SiP. The atomic structure oriented along the [310] zone axis is shown in Fig. S2(b). In this case also, one can notice the presence of stacking faults when following the A and B type layers. Sometimes, even three to four rows are found to have the same chevron orientation (see the sequence indicated in Fig. S2(a)).